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RESEARCH IN THE RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT

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Cincinnati, Ohio

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RESEARCH IN THE RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT

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U. S. Department of Health, Education, and Welfare

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RESEARCH IN THE RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT

ABSTRACT

Radioactivity may be found anywhere in the environment at the present time. In the past most of that encountered resulted from cosmic radiation and from the naturally occurring radionuclides. In any evaluation of the exposure of man to radiation, consideration must be given to cosmic rays and natural occurring radioactivity as well as to medical sources and man-made radioactivity released into the environment.

In this paper, the studies under way at the Robert A. Taft Sanitary Engineering Center, Cincinnati, to define the levels of radioactive contamination in environmental samples, are described. Currently, measurements of gross activity are being made along with a few measurements of the more hazardous radionuclides. In addition, some other nuclides are identified. The levels of activity in air (from fallout and natural radioactivity), in rainfall, snow, cistern and surface waters, and in milk and other food materials, are indicated. The contribution of these sources to the strontium-90 intake has been estimated and areas in which additional studies must be carried out delineated.

RESEARCH IN THE RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT

The sole sources of radioactive contamination of the environment prior to 1945 were cosmic radiation, which resulted in the formation of such radionuclides as H^3 and C^{14}, and the naturally occurring radionuclides, such as uranium, thorium, and their daughter products; potassium-40; rubidium-87; and several others. With the harnessing of nuclear energy, many additional sources of radioactive contaminants have become available including nuclear fuel fabrication facilities; reactor installations - both fixed and mobile; nuclear fuel reprocessing plants; beneficial applications of radioisotopes in medicine, research, and industry; and fallout from nuclear tests.

Release of radioactive materials from any of the operations indicated above can lead to the direct contamination of workers, the general population, or various segments of the environment. Although the responsibility for control of such pollutants is with the operator of the facility involved, local and national health agencies are necessarily interested in the wide-scale public health implications of such releases.

Exposure from these environmental sources must be considered along with exposures due to the use of radiation in medical diagnosis

and therapy, and to radiation from natural sources. Dr. Lauriston S. Taylor (1) and others (2, 3) have indicated that on the basis of genetic considerations the exposure to age 30 should not exceed 14 million rem per million population distributed as follows from each of these major sources:

Table 1

RADIATION DOSE RECEIVED UP TO AGE 30 (1)

(MPI = 14,000,000 man-rem per million persons)

Source	Dose
	(man-rems)
Natural radiation	4,000,000
Medical irradiation	5,000,000
Occupational Exposure	150,000
Radiation in plant environs	
At one-tenth of new MPD	(150,000)
At one-tenth of old MPD	(450,000)
Fallout	200,000
Total	9,800,000
Balance	4,200,000

This paper will describe the research activities of the Division of Radiological Health, Robert A. Taft Sanitary Engineering Center, Public Health Service, Cincinnati, to define the levels of natural radiation, radiation in plant environs (environmental contamination), and radiation due to fallout.

Approach

The determination of radioactive contamination of the environment is approached through two routes. The first is concerned with the measurement of gross alpha, beta, or gamma activity in specific media such as air, water and biological materials, and is of value to indicate the trends and changes taking place in the total radiation picture. Such measurements will serve also as a basis for routine control in nuclear energy establishments, to indicate accidental release of radioactivity in plant operations, to follow the movement of radioactive clouds formed during nuclear detonations held in the open, and to estimate the external dose contributed by such release.

Since gross radioactivity measurements cannot be directly related to hazards, particularly low level hazards, the second approach involves the identification of specific radionuclides in a wide variety of environmental media including surface, ground, and marine waters and their associated biota; bottom muds; air; milk and other

food materials; bone; vegetation; soils; etc. Although an extensive methodology for specific radionuclides exists, much of it has been developed for the identification of relatively high concentrations in specialized types of samples. It has been necessary, therefore, to modify these methods where this could be done or to develop new ones applicable to the low concentrations encountered in a wide variety of environmental media and in the presence of large amounts of stable materials and even of naturally occurring radioactivity. In all method developments the optimum combination of radiochemistry and instrumentation is employed.

Surveillance Networks

Although radiation surveillance activities are carried on by many agencies including the Atomic Energy Commission, the Department of Agriculture, the U. S. Weather Bureau, the Department of Defense, and the Department of Health, Education, and Welfare, only those operated through the various programs at the Robert A. Taft Sanitary Engineering Center will be reviewed here.

The Center operates three national networks in cooperation with state and municipal agencies. These include the National Air Sampling Network which comprises 161 stations from which 24-hour samples of air particulates are collected on a statistically random basis at least bi-weekly; the Water Quality Basic Data Network, which collects weekly samples from 50 stations on the principal U. S. streams;

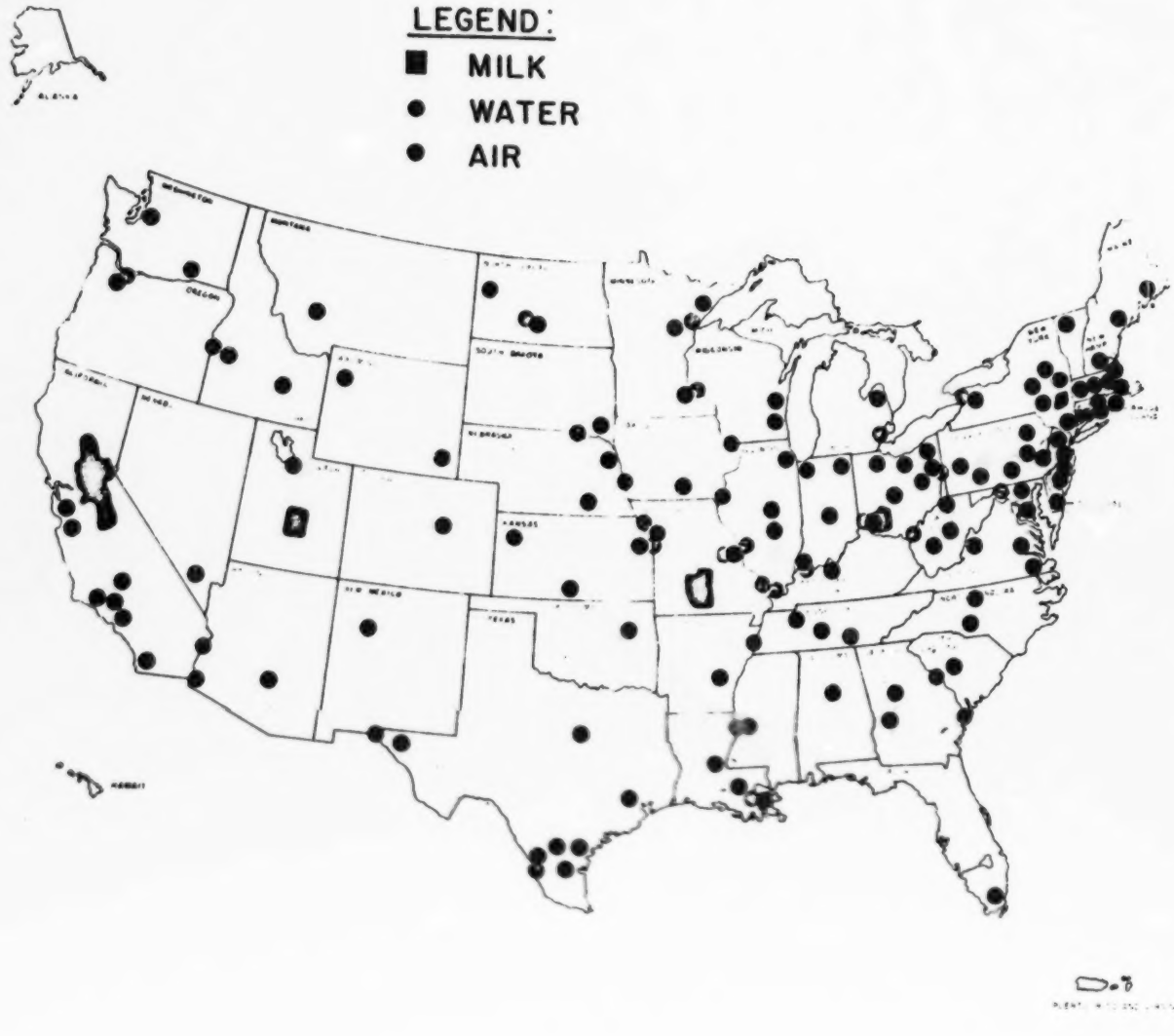
and a National Milk Surveillance Network which provides monthly samples from each of 10 milk sheds. Gross beta determinations are made on the air and stream samples; gross alpha on one-fourth of the stream samples; and specific radionuclide analyses on the milk samples. The location of all sampling stations, with the exception of 5 milk sheds, are indicated in Figure 1.

At Cincinnati gross and specific radionuclide levels are determined also in rainfall, cistern waters, vegetation, and various other food materials, as well as natural radon and thoron daughter products.

Air Samples

The 24-hour samples of suspended air particulate matter are collected on 8 x 10 inch glass fiber filters. Upon return to the Center, the samples are weighed and counted by placing the entire filter under a large proportional gas flow counting chamber (7-1/2 inches in diameter) having a thin metal-coated Mylar window. The piston of the counter chamber has been replaced with a steel plate, covered with aluminum foil for ease of decontamination which fixes the sample 5 mm below the thin-window. Figure 2 shows the filter being placed in the counter.

To allow decay of the natural alpha and associated beta-gamma activity, the samples are counted after two days and again after



ENVIRONMENTAL SAMPLING STATIONS FOR RADIATION SURVEILLANCE

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four to seven days to establish the decay in terms of apparent half-life and to estimate the time of formation of the mixture. From these measurements, the gross beta activity of air at the time of collection can be determined.

The data obtained for Cincinnati over the period 1953-1957 are summarized in Table 2. These data show that the air particulate radioactivity levels have increased from an average value of 0.2 $\mu\mu c/M^3$ in 1953 to 4.3 $\mu\mu c/M^3$ in 1957 (4).

Table 2

CINCINNATI, OHIO, AIR RADIOACTIVITY DATA - $\mu\mu c/M^3 \times 10^{-1}$ - 1953-1957 (4)

Year	No. of Samples	Min.	Max.	Avg.	Frequency Distribution - Per Cent								
					10	20	30	40	50	60	70	80	90
1953	24		6	2		1	1	2	2	3	4	4	5
1954	250		107	6		1	1	2	3	4	6	10	16
1955	55	1	64	11	1	1	2	3	6	9	15	19	30
1956	40	6	67	20	8	11	14	16	18	20	22	25	33
1957	18	5	219	43	9	15	18	23	28	31	38	42	88

In addition to the measurement of air particulate activity, some information has been collected on the levels of radon and thoron daughter activity in the vicinity of the Center. As expected, considerable diurnal variations are encountered along with climatological variations. Studies conducted during the past year show morning radon

activity levels varying from 70 to 2320 $\mu\mu c/M^3$. Measurement of afternoon samples was initiated in July, 1958, and during the six months' period while the morning radon levels ranged from 70 to 2100 $\mu\mu c/M^3$, the afternoon levels ranged from 8 to 400 $\mu\mu c/M^3$. Thoron levels ranged from 4 to 10 with an average of 7 $\mu\mu c/M^3$ in October and from 1 to 5 $\mu\mu c/M^3$ in November.

Earlier radon levels were summarized (see Table 3) on the basis of calm vs. wind conditions (calm-wind less than 2 mph; windy-wind greater than 2 mph) and rain under the same conditions of calm and wind.

Table 3

AVERAGE RADON LEVELS - CINCINNATI AIR

Condition	Activit · $\mu\mu c/M^3$
Calm (< 2 mph wind)	1850
Calm (> 2 mph wind)	535
Rain (< 2 mph wind)	690
Rain (> 2 mph wind)	495

Radon levels were determined in the pipe tunnel at the Center and the levels were surprisingly high as indicated in Table 4, and it may be worthwhile to check basement and pipe tunnel levels in other Cincinnati buildings. Levels would depend on location, geologic formations, building materials, ventilation provided, etc.

Table 4

RADON LEVELS - SEC PIPE TUNNEL - $\mu\mu c/M^3$

Date	Time	Activity	Date	Time	Activity
11/ 5/58	8 a.m.	883	11/20	4 p.m.	837
11/14	4 p.m.	4230	11/24	4 p.m.	1886
11/17	5 p.m.	1185	12/ 3	10 a.m.	732
11/18	9 a.m.	834			

Water

Under this category will be summarized the results obtained in sampling rainfall, snow, cisterns, and surface waters.

Rainfall is collected in a rectangular container two square meters in area. Aliquots of the material are passed through millipore filters for the separation of radioactivity associated with the suspended and dissolved fractions of the sample and counted. Rainfall levels reflect nuclear test operations and, in general, a larger percentage of activity is found in the suspended fraction shortly after such tests. There is a gradual decrease in the fraction of the radioactivity associated with suspended solids. In Table 5, the values found in rainfall, snow, cistern and surface waters during the past year are indicated.

Table 5

RAINFALL, SNOW, CISTERN AND SURFACE WATER GROSS BETA RADIOACTIVITY LEVELS - CINCINNATI

Month	Rainfall		Snow		Cistern Waters		Surface Waters	
	No. of Samples	Radioactivity μμc/l	No. of Samples	Radioactivity μμc/l	No. of Samples	Radioactivity μμc/l	No. of Samples	Radioactivity μμc/l
1958								
Jan.	4	390-1,712	4	230-560			4	7-108
Feb.	3	500-910	5	140-2,255	3	105-134	4	1-49
Mar.	7	442-9,840	4	1,081-2,187	8	126-197	5	4-70
Apr.	11	656-15,047			13	138-285	3	31-247
May	9	912-9,901			13	175-297	4	27-61
June	8	1,328-8,476			13	102-317	5	1-60
July	7	3,003-6,022			15	225-512	4	39-340
Aug.	10	834-5,381			6	389-510	4	42-141
Sept.	7	258-9,811			10	116-405	5	1-39
Oct.	4	451-3,325			8	205-317	5	7-36
Nov.	11	1,313-21,924			10	288-480	4	28-59
Dec.					9	294-396	5	25-164

Rainout data in terms of curies per square mile for the period 1953-1957 are shown in Figure 3 and indicate that during this period the levels increased from less than 0.1 to about 0.54 curies per square mile.

Gamma spectral measurements and isotopic analyses, particularly for the determination of total radio-strontium and strontium-90 have been made. The isotopes identified from the gamma spectral measurements include Ce^{141} and/or Ce^{144}, I^{131}, Ru^{103} and/or Ru^{106}, Cs^{134}, Cs^{137}, Zr^{95}-Nb^{95}, Zn^{65}, Co^{60}, and K^{40}. The strontium levels found in some rainfall samples are summarized in Table 6 and range from 0.6 to 12.3 μμc/l for total strontium and from 0.3 to 7.5 μμc/l for Sr^{90}.

Table 6

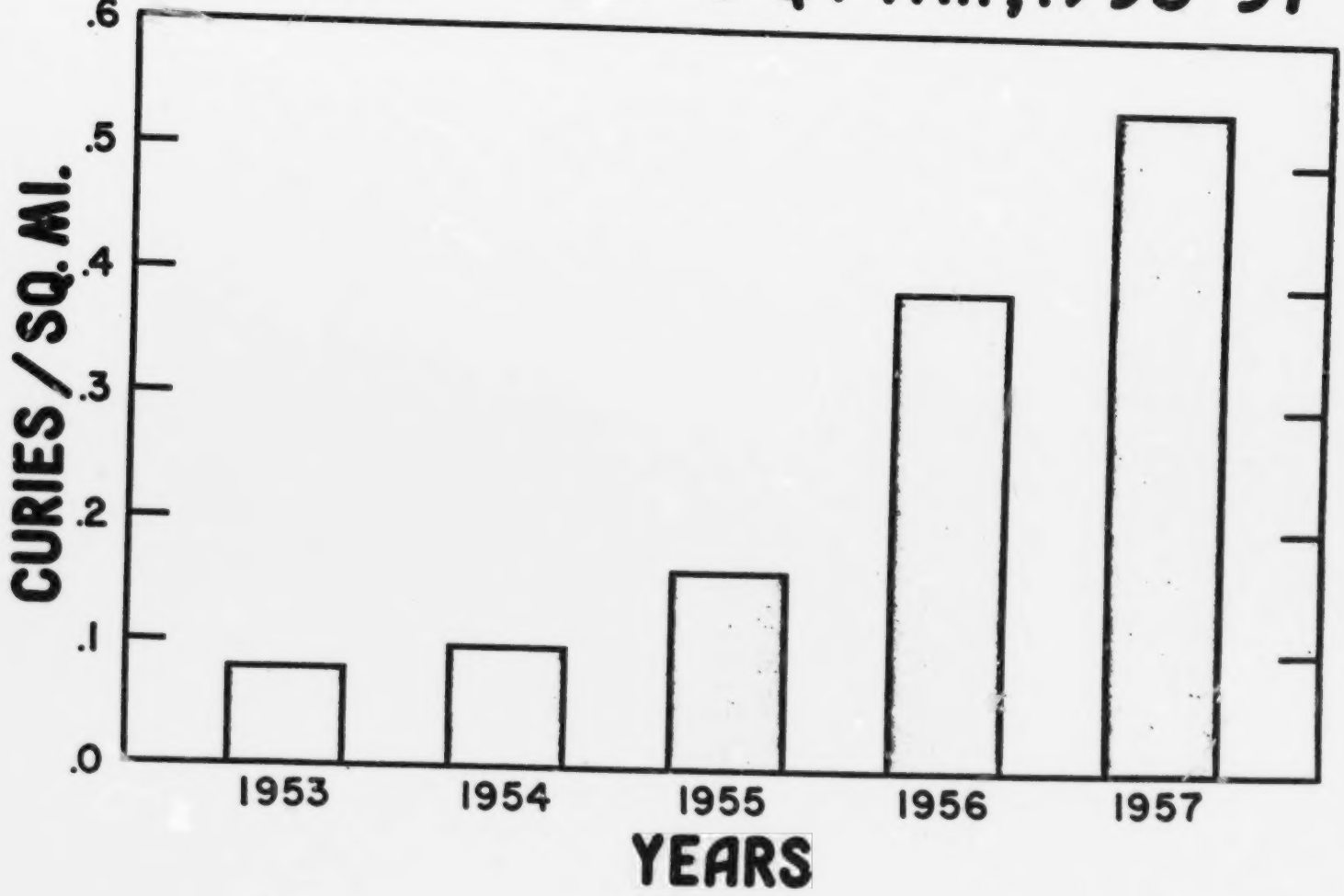
RADIOACTIVE STRONTIUM IN CINCINNATI RAINS - μμc/l*

Date	Total Strontium	Strontium-90	Remarks
6/14/57	5.5 ± 0.6	4.9 ± 0.4	
6/17/57	12.3 ± 0.5	7.5 ± 0.4	
6/24/57	0.6 ± 0.05	0.3 ± 0.04	
6/27-28/57	2.1 ± 0.1	1.3 ± 0.1	Dissolved fraction
	0.7 ± 0.1	0.9 ± 0.1	Suspended fraction
7/4/57	1.8 ± 0.1	1.3 ± 0.06	
9/9/57	4.6 ± 1.1	3.6 ± 1.1	

* Errors are at 95 per cent confidence level.

RAINOUT IN CINCINNATI

CURIES PER SQ. MI., 1953-57



Since rainfall serves as a more direct source of water supply where cisterns are used, radioactivity levels were determined for such a supply. The activity levels have increased during the year from an average value of 119 μμc/l in January to a value of 346 μμc/l in December. The highest activity levels were found in August with an average of 459 μμc/l for six determinations. Gross radioactivity data have been summarized in Table 5. Gamma scans show the presence of Ce^{141} and/or Ce^{144}, Ru^{103} and/or Ru^{106}, Zr^{95}-Nb^{95} and K^{40}. Samples are currently being composited on a quarterly basis for the determination of total strontium and strontium-90.

Radiochemical analyses were performed on cistern sludge samples collected in March, 1955, and March-May, 1956. The strontium levels found are in shown in Table 7.

Table 7

STRONTIUM LEVELS IN CINCINNATI CISTERN SLUDGES

Date Collected	Radioactivity - μμc/g. ash	
	Total Sr	Sr^{90}
March, 1955	1.0	n.d.
March, 1956	1.4 - 9.2	0.4 - 3.1
May, 1956	4.4	0.3 - 0.6

Qualitative gamma spectral measurements were also made on two of these sludges with the results indicated in Table 8.

Table 8

GAMMA RADIOACTIVITY - CINCINNATI CISTERN SLUDGE

Date Collected	Peak Radioactivity - cpm/50 kev			
	Ce^{144}	Ru^{106}	Cs^{137}	Zr^{95}-Nb^{95}
March, 1955	71	16	50	--
March, 1956	254	52	152	13

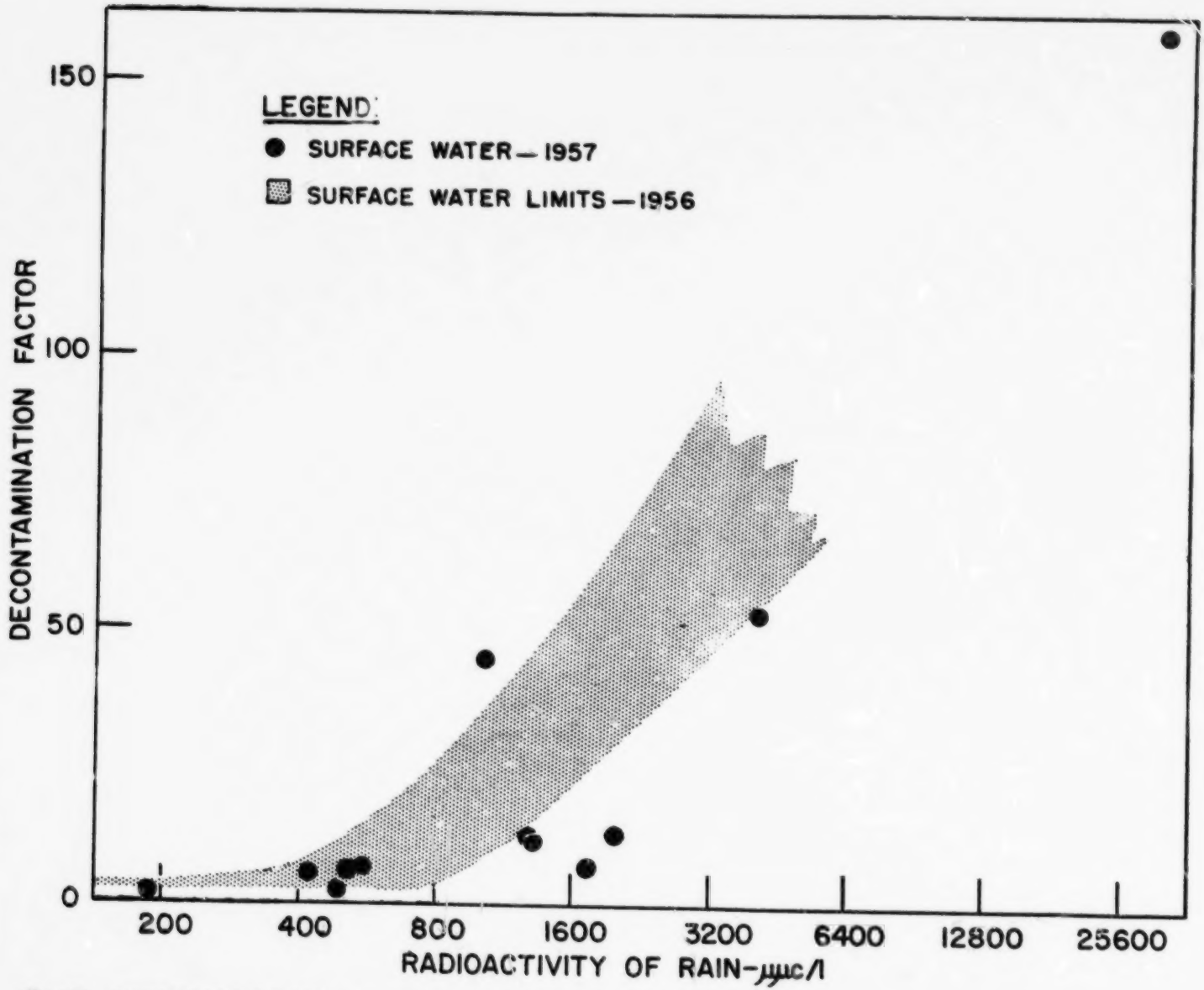
In connection with the Water Quality Basic Data program, samples of Ohio River water have been collected at Cincinnati and analyzed for gross radioactivity. The levels, which are summarized in Table 5, show variations from 1 to 340 μμc/l for gross radioactivity. Here again, in processing the samples, the gross radioactivity associated with the suspended and dissolved fractions is determined separately. Radioactivity associated with the suspended fraction is of special interest where the water treatment process incorporates chemical treatment, sedimentation and filtration. During such treatment the bulk of the radioactivity associated with suspended matter should be removed and should not reach the consumer. In addition, depending on the chemical treatment used, some of the radioactivity associated with the dissolved fraction will be removed (5, 6). Approximately 44 per cent of the radioactivity was associated with the suspended fraction.

The ratio of total rain activity to total surface water activity was determined for the several months in which comparable data were available and reported in terms of the decontamination factor (rainout or fallout activity divided by the surface water activity). These decontamination values were plotted against the rain activity as shown in Figure 4 (7, 8). It will be seen that removal of radioactivity by natural agents increases with increased radioactivity of the rain.

Milk and Other Foods

Measurement of radioactivity levels in Cincinnati milk was initiated in May, 1957, on a monthly schedule. The selection of sampling stations within the milk sheds was based upon criteria (9) to assure that the milk represented at least 1,000 cows in a metropolitan milk shed and that each monthly sample was representative of the same farms. In addition, the number of farms represented had to be small enough to permit the collection of collateral field data, including feeding practices, water supplies, and cattle breeds.

All milk samples received have been analyzed routinely for iodine-131, strontium-90, strontium-89, barium-140, cesium-137, and potassium-40,



THE DECONTAMINATION OF RAIN BY NATURAL AGENCIES

the isotopes contributing most of the activity, and for stable calcium. Upon receipt at the Center, the fluid milk is placed in specially designed beakers for counting in the 100-channel gamma spectrometer. From this count, the iodine-131 content is determined. The fluid milk is ashed, the total radioactive strontium, strontium-90, barium-140, and cesium-137 obtained by radiochemical methods, and the stable calcium and potassium are determined by flame spectrophotometry. The data obtained in 1958 are indicated in Table 9. Alpha counting of the barium precipitate showed that the milk contained from 0 to 0.3 μμc/l of alpha activity, which presumably is radium.

Typical food materials comprising specific dietary components were purchased on the Cincinnati market and analyzed for gross beta-gamma activity, strontium-90, and stable calcium. The results of these studies are summarized in Table 10.

Summary

In the previous sections of this paper, levels of radioactivity in various environmental media were indicated. The values are recapitulated below (Table 11), particularly in terms of the strontium-90 levels found. This has been done to try to estimate the intake of this radionuclide from the sources indicated. From the estimates made, it can be seen that of the four environmental media considered, air contributes the least amount of strontium with water (either cistern or surface), milk and food all contributing about equal amounts on the average. In the case of

Table 9

RADIOACTIVITY DATA - CINCINNATI MILK - 1958

Month	Radioactivity - μμc/l					Stable Calcium
	Sr^{90}	Sr^{89}	Ba^{140}	I^{131}	Cs^{137}	grams/liter
January	4.1	0.3	0	0	48.3	1.180
February	3.9	8.5	1.7	0	55.5	1.154
March	4.1	5.6	1.7	0	61.6	1.080
April	6.4	13.1	0	30	50.2	1.144
May	8.6	70.4	15.6	0	50.1	1.161
June	11.6	106.9	35.7	0	77.5	1.227
July	13.0	193.9	112.1	107	95.8	1.082
August	7.5	96.9	83.8	42	84.2	1.102
September	9.4	97.8	13.4	5	58.6	1.132
October	8.6	56.2	7.8	31	53.1	1.175
November	15.2	108.0	10.3	177	63.6	1.157
December	9.6	37.0	4.5	8	62.8	1.148
Average	8.5	66.2	23.9	33	63.4	1.145

Table 10

RADIOACTIVITY LEVELS IN CINCINNATI MARKET FOODS

Sample	Radioactivity - μμc/kg		Stable Calcium
	Gross β-γ	Sr^{90}	gm/kg
Eggs	1,360	3.0	0.695
Fruits and juices	1,780-2,190	2.1-3.7	0.128-0.178
Pastries and cereals	1,180-2,540	10.6-12.4	0.428-0.480
Rice	740	1.2	0.196
Dairy products	540	39.0	4.836
Seafoods and meats	2,310-2,520	0.2-1.0	0.172-0.335
Vegetables	2,840-3,240	0.234-0.508	3.9-12.5
Beverages (Instant)	40,100-45,300	19.0-60.0	1.900
Mixed nuts	6,200	8.1	0.685
Milk		8.5	1.145

Table 11

RADIOACTIVE CONTAMINATION - ENVIRONMENTAL SOURCES

Source	Radionuclide Activity Level - μμc/l, μμc/M³, μμc/kg	Daily Intake ml or grams	Intake of Radionuclide μμc/day	Source of Data
Air	Sr^{90} - 10^{-4}-1.2x10^{-2} μμc/M³	2 x 10^7 ml	0.002 - 0.24	(10)
Water - Cistern or Surface	Sr^{90} - 0.3-7.5 μμc/l	2,200 ml	0.7 - 16.5	
	Sr^{90} - 1-5 μμc/l	2,200 ml	2.2 - 11	
Milk	Sr^{90} - 8.5 μμc/l	1,000 ml	8.5	(9)
Food	Sr^{90} - 0.2-60 μμc/kg	~1,200 gm	?	(11)
	Totals		[16.2-17.7] min.	
			[26.2-32.2] max.	
	% of permissible intake (based on water)		(9-10)-(15-19) %	
Air	Radon - 8.3-2,100 μμc/M³	2 x 10^7 ml	166 - 42,000	
Air	Thoron - 4-10 μμc/M³	2 x 10^7 ml	80 - 200	

food materials, the intake was estimated in terms of the individual strontium values given in Table 10 and the estimated daily intake of the various food materials in the diet. The value given for food intake is exclusive of milk, but not exclusive of dairy products. Comparing the total daily intake of strontium-90 to the permissible intake for water (80 μμc/l x 2.2 liters/day = 176 μμc/day) it will be seen that the intake represents 9 to 19 per cent of the allowable daily intake of this radionuclide. This, of course, does not represent the total exposure, and similar calculations must be carried out based on the intake of other radionuclides. Because of their biochemical and physical properties, most of the other radionuclides are much less hazardous than strontium-90 so that the estimation of the strontium-90 exposure accounts for the greater part of the problem. Since radon and thoron levels have been reported earlier, estimates have been made of the total intake of these naturally occurring radionuclides by inhalation.

Conclusions

Although this paper essentially reports radioactivity levels in Cincinnati samples, radioactivity is being determined in many areas on a wide variety of samples. The work is by no means complete, and more studies are needed to define the current levels of specific radionuclides in environmental media. Those areas showing higher levels should be studied in greater detail to determine the sources of these higher levels and also to follow the movement of the radionuclides of interest through the various segments of the environment and the food chain to man. With such information at hand remedial measures may be instituted to minimize the transfer

of radioactivity through the environmental cycle to man. It is not too soon to plan and carry out studies which will provide means for controlling the spread of specific radionuclides directly to man or through his food chain.

Information on environmental sources of radioactivity is needed to provide a better basis for indicating permissible occupational exposures. If environmental sources result in significant exposure to the individual, it will be necessary to reduce occupational levels to a point where hazard will be minimized. Furthermore, exposure from occupational sources may be controlled more easily than exposure from environmental sources.

Acknowledgments

The data reported herein were obtained by personnel in the Radiological Health Research Activities Unit and the Milk and Food Program, Robert A. Taft Sanitary Engineering Center. It is with great sincerity that I acknowledge use of these data.

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